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D2.1 Pollutant:CO_2 Ratio Analysis

Final Report

Breathe London Project

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CERC

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Author(s): Ella Forsyth

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1. Introduction

The high time resolution (1 second frequency) of the Breathe London mobile measurements offers the opportunity to distinguish locally emitted components from other components of the total concentration at a measurement location since these show the highest frequency variation. The value of the ratio of local NO_X and local $PM_{2.5}$ to local CO_2 gives an indication of the type of engine that produced the emissions. These ratios can then be compared with equivalent ratios in the emissions inventory typically used for modelling air quality in London, to provide insight into the accuracy of both the emission factors and the relative numbers of different vehicle types used in compiling the inventory. This short report provides a summary of the work done by CERC to compare NO_X:CO_2 and $PM_{2.5}$:CO_2 ratios calculated from the hyperlocal component of the mobile measurements with those calculated from the emissions inventory used in Breathe London modelling. Section 2 describes the assessment methodology; Section 3 contains the overall results and Section 4 discusses the possible explanations and responses.

2. Methodology

2.1 Emissions

Emissions of NO_X and $PM_{2.5}$ from road traffic were obtained using annual average traffic flows and speeds from the London Atmospheric Emissions Inventory (LAEI) 2013 dataset, interpolated to 2019 between the 2013 base year and 2020 future predictions, combined with road traffic emissions factors from the Emission Factor Toolkit (EFT) v8 for 2019. The future predictions in the LAEI 2013 dataset incorporated the TfL Business plan, which included the planned introduction of the Ultra Low Emission Zone (ULEZ) in 2020 in Central London. The ULEZ was introduced on the 8th April 2019, one year earlier than planned, and any additional fleet changes that are attributed to this are not accounted for in the emissions. The emissions for each major road contained "real world" adjustments[1] for NO_X, and included the updates from the conclusions of the Hotspot Analysis report D3.2. The emissions for each road in the inventory were matched to a 30m road segment, with a limiting distance of 12m to ensure the correct data points were assigned to the nearest road. Data points were excluded from the comparison if there were no emissions inventory roads within this distance. The emission rates for CO_2 were calculated using COPERT emission factors[2].

2.1.1 Time-varying emissions profiles

Time-varying emissions profiles that represent differences in road traffic fleet composition were applied to the annual average emissions for each road. Each road in the inventory was classified by location (Central, Inner, Outer and Motorway) and type (A Road Single or Dual Carriageway, B Road, Minor Road, Local Street, Motorway). Not all types are in every location, so there are 17 road categories overall. All roads in each category were grouped together, and average flows for each road category in terms of 11 vehicle categories were calculated. DFT raw traffic flow data for London in 2018 were used to derive diurnal flow profiles for each vehicle category (DfT data only available weekdays between 07:00 and 18:00). These traffic flow diurnal profiles were then applied to the average emissions from each vehicle category to develop pollutant-dependent emissions profiles for each of the 17 different road categories for a sample weekday. Saturday and Sunday use the same standard hourly variation, as there is no DfT data available to refine it. Diurnal emissions profiles were developed in this way for each of the 17 road categories for NO_X, $PM_{2.5}$ and CO_2.

To represent traffic flow differences between different days of the week, hourly NO_X concentrations at kerbside reference monitors were used as a proxy for traffic flow; these were averaged over each day of the week from 1st October 2018 to 29th February 2020 (avoiding the period affected by COVID-19) to calculate the average concentration distribution by day of the week. This provided seven daily factors, which were then applied to the diurnal profiles, resulting in an individual diurnal profile for each day of the week for each road category (Figure 2-1). Representing the traffic flow in this way enables the higher traffic flow on weekdays and lower traffic flows over the weekend to be better represented.

[1] Factor calculations for real world adjustments done by CERC based on the initial work done by: Carslaw, D and Rhys-Tyler, G 2013: New insights from comprehensive on-road measurements of NOx, NO2 and NH3 from vehicle emission remote sensing in London, UK. Atmos. Env. 81 pp 339–347.

[2] COPERT Emission factors extracted from Annex 3 of chapter '1.A.3.b.i-iv Road transport' in the EMEP/EEA air pollutant emission inventory guidebook 2016: http://www.eea.europa.eu/publications/emep-eea-guidebook-2016

Emissions ratios were calculated for each 30m road segment by multiplying the annual average emission rates of NO_X, $PM_{2.5}$ and CO_2 by the appropriate hourly factor from the 7-day diurnal emissions factors (depending on the road category) for every hour in a representative 7-day period. This resulted in 168 (7x24) hourly NO_X:CO_2 and $PM_{2.5}$:CO_2 emissions ratios for each road segment, for comparison with the corresponding hourly measured ratios.

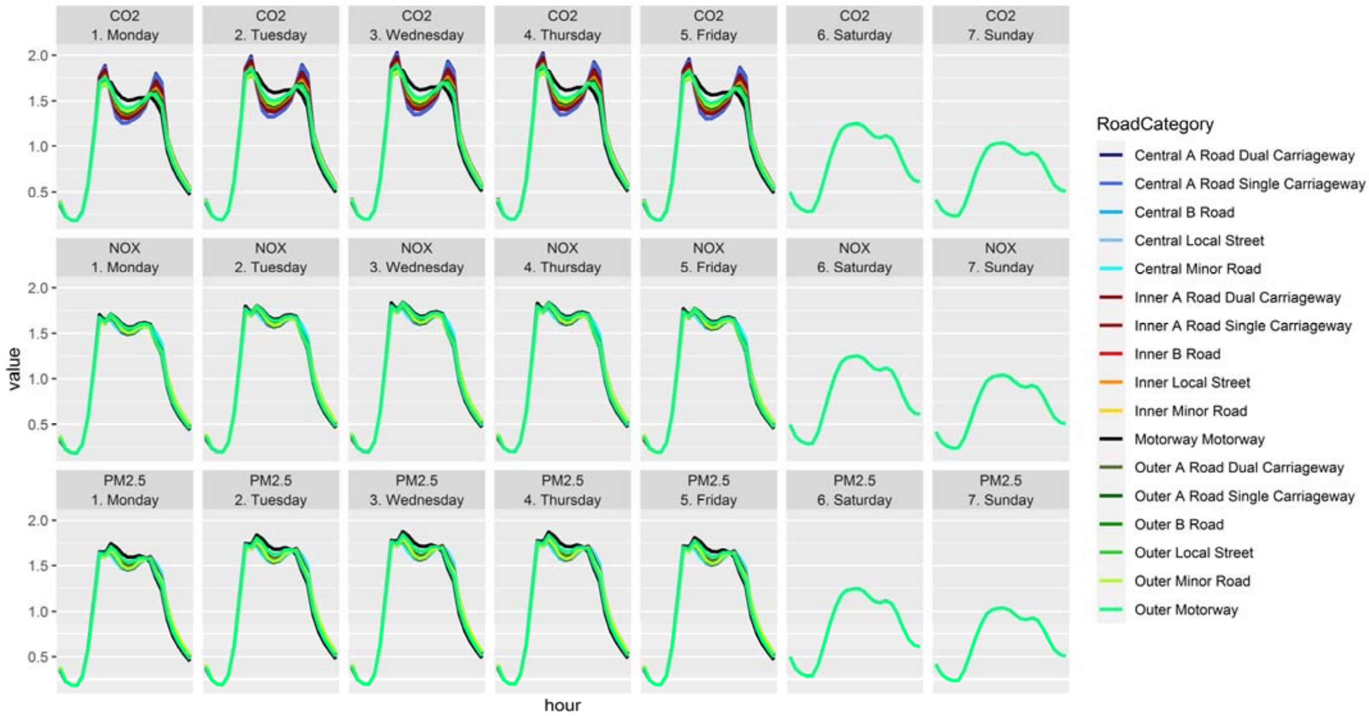

Figure 2-1 Pollutant dependent diurnal emissions profiles for each day of the week, for each road category

2.1.2 Units considerations

Road traffic emissions are in units of g km^{-1} s^{-1} for all pollutants and must be converted to ppb:ppm and µg/m^3:ppm for NO_X:CO_2 and $PM_{2.5}$:CO_2 ratios respectively, to match the units in the measured data.

$$\frac{NO_X[ppb]}{CO_2[ppm]} = \frac{C_{NOX} \times NO_X[g\ km^{-1}\ s^{-1}]}{\frac{C_{CO2}}{1000} \times CO_2[g\ km^{-1}\ s^{-1}]}$$

$$\frac{PM_{2.5}[\mu g\ m^{-3}]}{CO_2[ppm]} = \frac{PM_{2.5}[g\ km^{-1}\ s^{-1}]}{\frac{C_{CO2}}{1000} \times CO_2[g\ km^{-1}\ s^{-1}]}$$

$C_{NOX} = 0.52$ and $C_{CO2} = 0.55$ are the conversion factors from µg/m^3 to ppb for NO_X and CO_2 concentrations respectively (NO_X emissions use the "NO_X as NO2" convention).

2.2 Measured data

The measured ratios were downloaded from the aggregated 30m ratio dataset (QAQC version 8) in the Street View Air Quality London data store in Google Big Query. The datasets contained the median of all valid 1-second ratios, calculated from all valid 1-second measurements from 2 cars along a 30m road segment, for each 1-hour time window the cars were driving. To isolate the hyperlocal component of the mobile data, a non-hyperlocal "baseline" was identified as the 1st percentile of concentrations within rolling 5 minute windows where windows were centered around each 1-second mobile measurement and removed from the measurements. The ratios were calculated with the remaining local enhancement component and then the median was calculated for each hour. The sensors did not measure NO_X concentrations directly, so NO_X was retrospectively calculated from NO and NO_2 in the quality controlled 1-second dataset.

The measured ratio dataset and the emissions ratio dataset were joined by the hour, day of the week and the individual 30m road segment, and then only the hourly measurements for hours in which there are valid Google car measurements during the driven period from 1st September 2018 to 31st October 2019 are retained for the comparison. The data was split by whether the road segment was inside or outside of the Ultra Low Emission Zone (ULEZ), and by whether the drive occurred before or after the 8th April 2019, when the Ultra Low Emission Zone was first introduced.

3. Results

This section describes the results comparing NO_x:CO_2 and $PM_{2.5}$:CO_2 ratios between the measured data and emissions.

3.1 NO_X:CO_2

Figure 3-1 is a histogram comparing the distribution of the NO_X:CO_2 ratios found in the emissions with the measurements. To better visualise the distribution, the ratios calculated from the measured data have been capped at 15. The data is filtered by whether the drive is before or after the introduction of the ULEZ and whether the road segment is located inside the ULEZ boundary. Overall, there is greater variation in the measurements, which is to be expected as these are influenced by local traffic conditions during the drive; the emissions inventory represents the average mix of vehicle types on each road. The most significant variation can be seen in b) for the roads outside of Central London that are driven before the introduction of the ULEZ on the 8th April 2019; there is much less variation in c) for roads inside the ULEZ after the introduction of the ULEZ. The reduction in the tail in the measured ratios suggests there was a greater proportion of cleaner vehicles across London after the introduction of the ULEZ.

Figure 3-2 is a box and whisker plot comparing the ratios in the measured data and emissions; the scale on the y-axis is truncated to only include ratios below 15. The median measured ratio is consistent with the emissions inventory equivalent, which suggests that adjusting the average NO_X emission rates provided by the emissions inventory by real world factors is correct, which is consistent with previous findings[3]. The medians show good agreement for the period after the ULEZ was introduced, both inside and outside the ULEZ, as emphasized in Table 3-1, with less good agreement before the introduction of the ULEZ, particularly

[3]Carslaw, D and Rhys-Tyler, G 2013: New insights from comprehensive on-road measurements of NOx, NO2 and NH3 from vehicle emission remote sensing in London, UK. Atmos. Env. 81 pp 339–347.

inside the ULEZ. If the same roads had been driven at the same time of day and week before and after the introduction of the ULEZ then there would be no change in the emissions inventory ratios pre- and post-ULEZ, therefore differences reveal the effect of the drive time of day and route on the results, which is largest inside the ULEZ. The difference in measured ratios pre- and post-ULEZ is much larger, suggesting that the majority of the reduction in measured ratios is due to traffic fleet changes. The differences between inside and outside the ULEZ are greater in the emissions inventory ratios than in the measured ratios, suggesting that the dominant effect observed in the measurements is the renewal of the traffic fleet with cleaner technology across London, not just inside the ULEZ.

Figure 3-3 shows how the monthly median ratio varies in the emissions and the measured data from the 1st September 2018 to 31st October 2019. There is a noticeable decline in the measured ratios over the first six months, both inside and outside the ULEZ, before plateauing after April 2019.

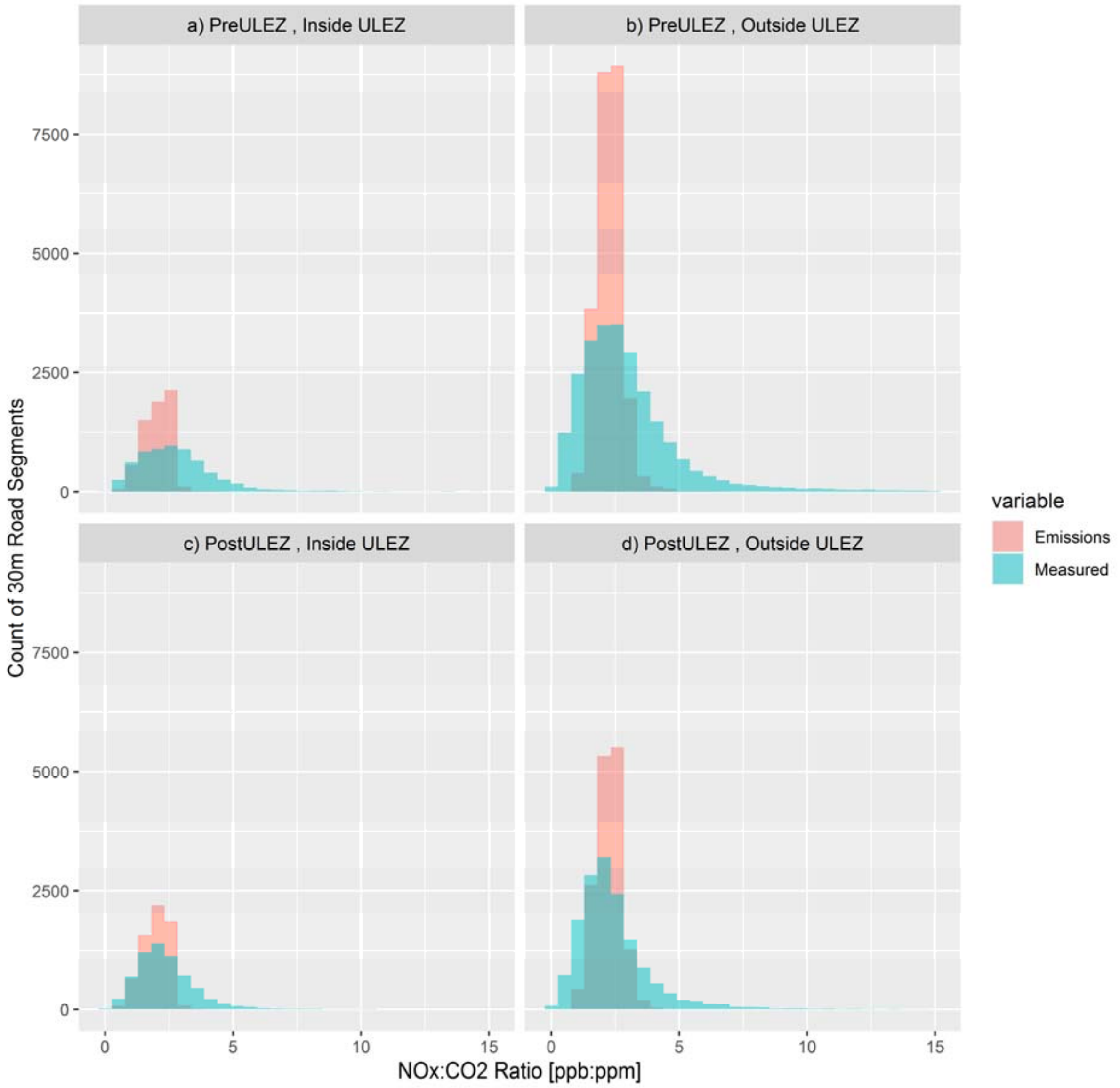

Figure 3-1: Histogram of NOx:CO2(ppb:ppm) ratios in emissions and measured data per 30m road segment, separated by before and after the introduction of the ULEZ and whether the road segment was inside or outside the ULEZ. This dataset has been capped at a ratio value of 15.

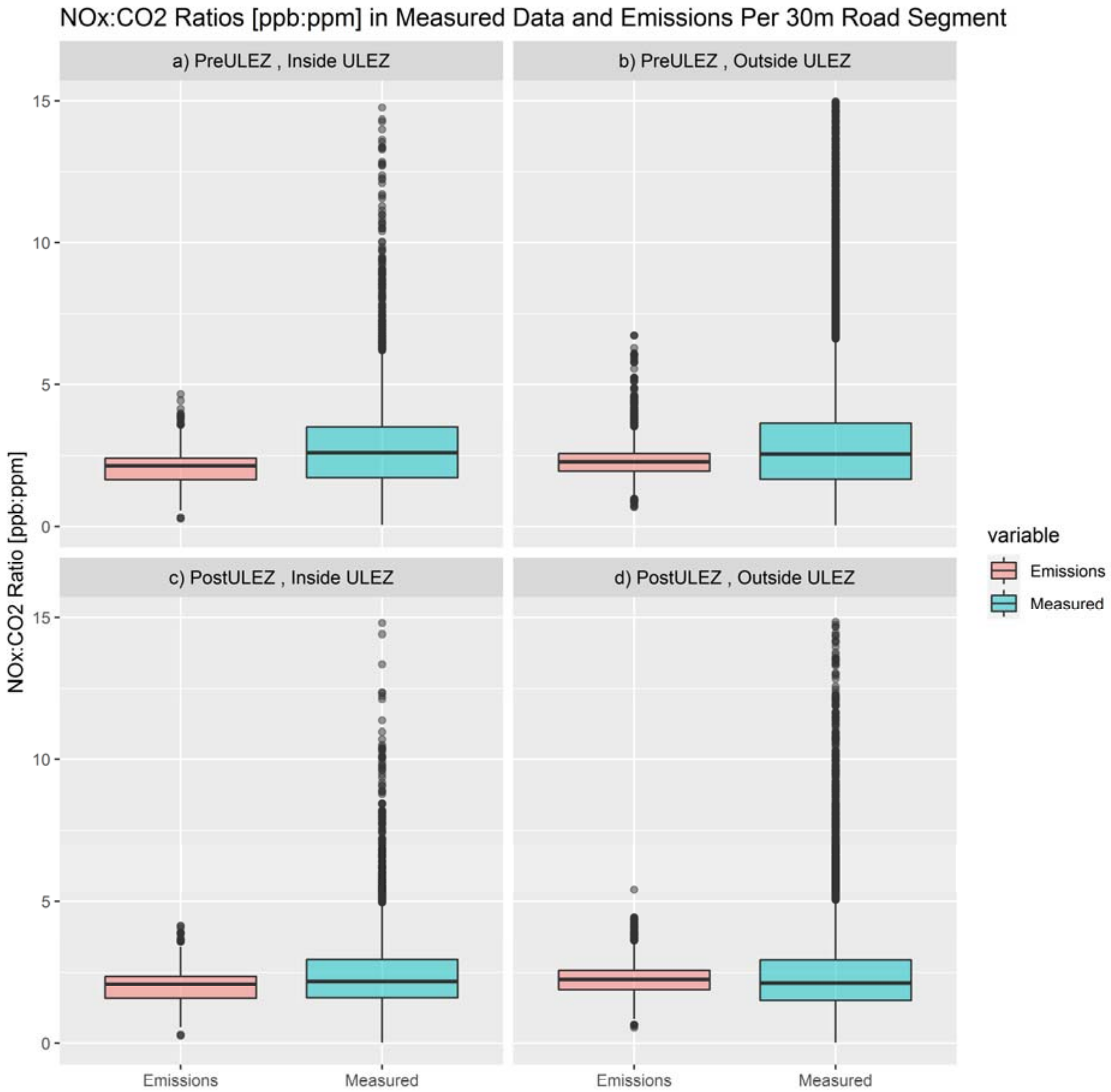

Figure 3-2: Boxplot of NOx:CO2 (ppb:ppm) ratios in emissions and measured data per 30m road segment, separated before and after the introduction of the ULEZ, and whether the road segment was inside or outside the ULEZ. The black dots represent the outliers This dataset has been capped at a ratio of 15.

Table 3-1 Table of the median NOx:CO2 (ppb:ppm) ratio in emissions and measured data per 30m road segment

		Emissions	Measured
Pre ULEZ	Inside ULEZ	2.14	2.61
Pre ULEZ	Outside ULEZ	2.29	2.57
Post ULEZ	Inside ULEZ	2.09	2.19
Post ULEZ	Outside ULEZ	2.26	2.14

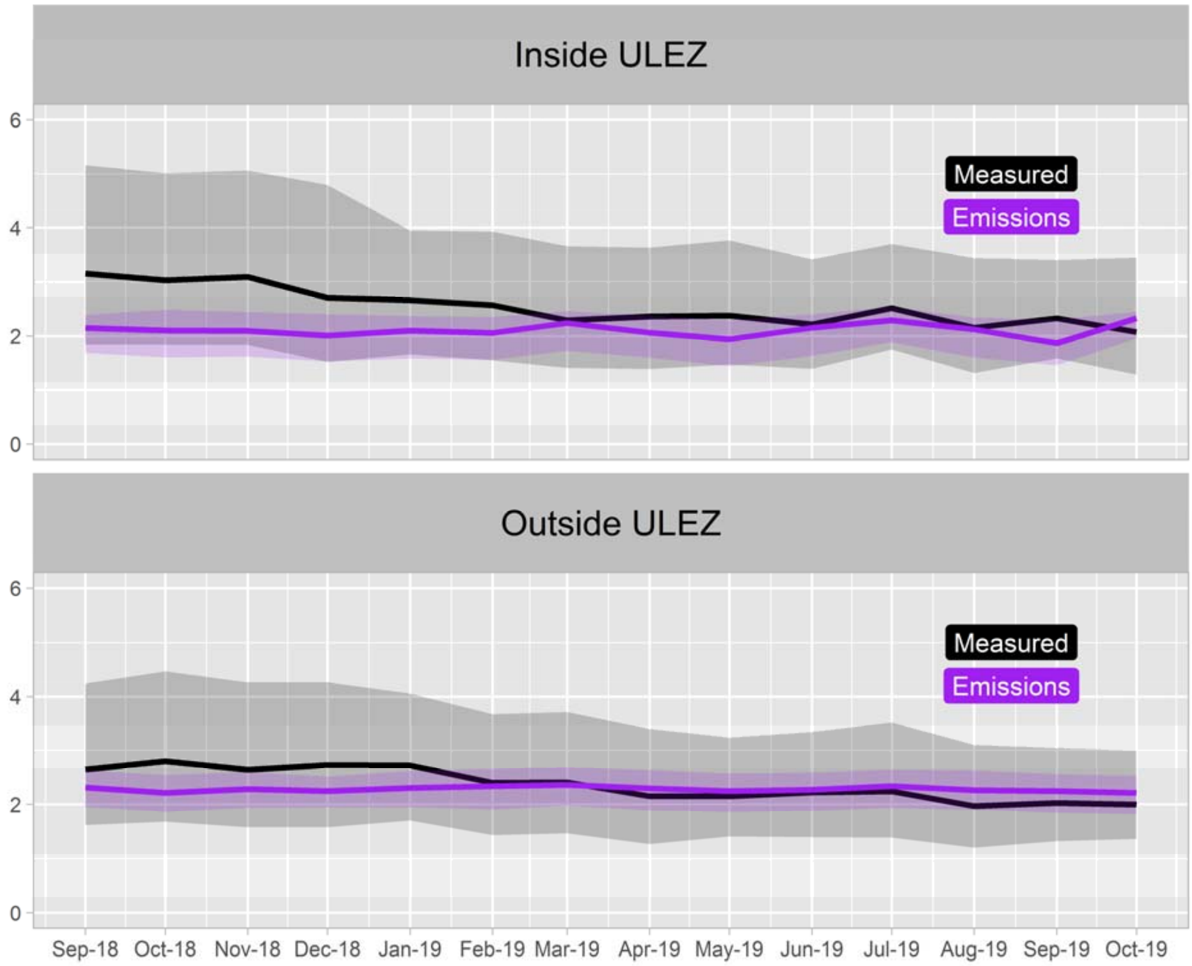

Figure 3-3: A time series of monthly averaged NOx:CO2 (ppb:ppm) ratios in measured data (black) and emissions (purple) for the period from the 1st September 2018 to 31st October 2019, separated by roads inside and outside the ULEZ. The bold line represents the median ratio and the faded lines represent the inter-quartile range

3.2 $PM_{2.5}:CO_2$

Figure 3-4 is a histogram comparing the distribution of the $PM_{2.5}:CO_2$ (ugm3:ppm) ratios found in the emissions with the measurements. To better visualise the distribution, the ratios calculated from the measured data have been capped at 5. The data is filtered by whether the drive is before or after the introduction of the ULEZ and whether the road segment is located inside the ULEZ boundary. Overall, there is greater variation in the measurements, which is to be expected as these are influenced by local traffic conditions during the drive; the emissions inventory represents the average mix of vehicle types on each road. There is minimal variation in the ratios in the emissions within each category.

Figure 3-5 is a box and whisker plot comparing the ratios in the measured data and emissions; the scale on the y-axis is truncated to only include ratios below 5. Table 3-2 shows the exact median values for each category and emphasizes that the ratio values for the emissions are significantly lower than the measured equivalent. For ratios inside the ULEZ, after the ULEZ was introduced the emissions (0.12) are 76% less than the measured equivalent (0.52). Across all categories the measured ratio median values are significantly higher than those in the emissions inventory, which suggests that the average $PM_{2.5}$ emission rates provided by the emissions inventory are too low, which is consistent with previous findings[4] that emissions inventories tend to underestimate the contribution from non-exhaust factors such as brake and tyre wear. There is more variation in the measured data with a larger interquartile range and a greater number of outliers. Similarly, there is a significant increase in variation in the measured data for the period after the ULEZ was introduced in April 2019.

Figure 3-6 shows how the monthly median ratio varies in the emissions inventory and the measured data from the 1st September 2018 to 31st October 2019. Unlike NOx, the ratios do not change significantly between September 2018 and April 2019. The medians surpass the median emissions ratios from April 2019 through October 2019, with a sharper increase for roads within the ULEZ. This increase could be explained by a large proportion of $PM_{2.5}$ emissions being due to non-exhaust emissions, which are largely unaffected by the renewal of the vehicle fleet with cleaner exhaust emissions technology, whereas CO_2 emissions are generally lower from newer vehicles, leading to an overall increase in the $PM_{2.5}$:CO2 ratios.

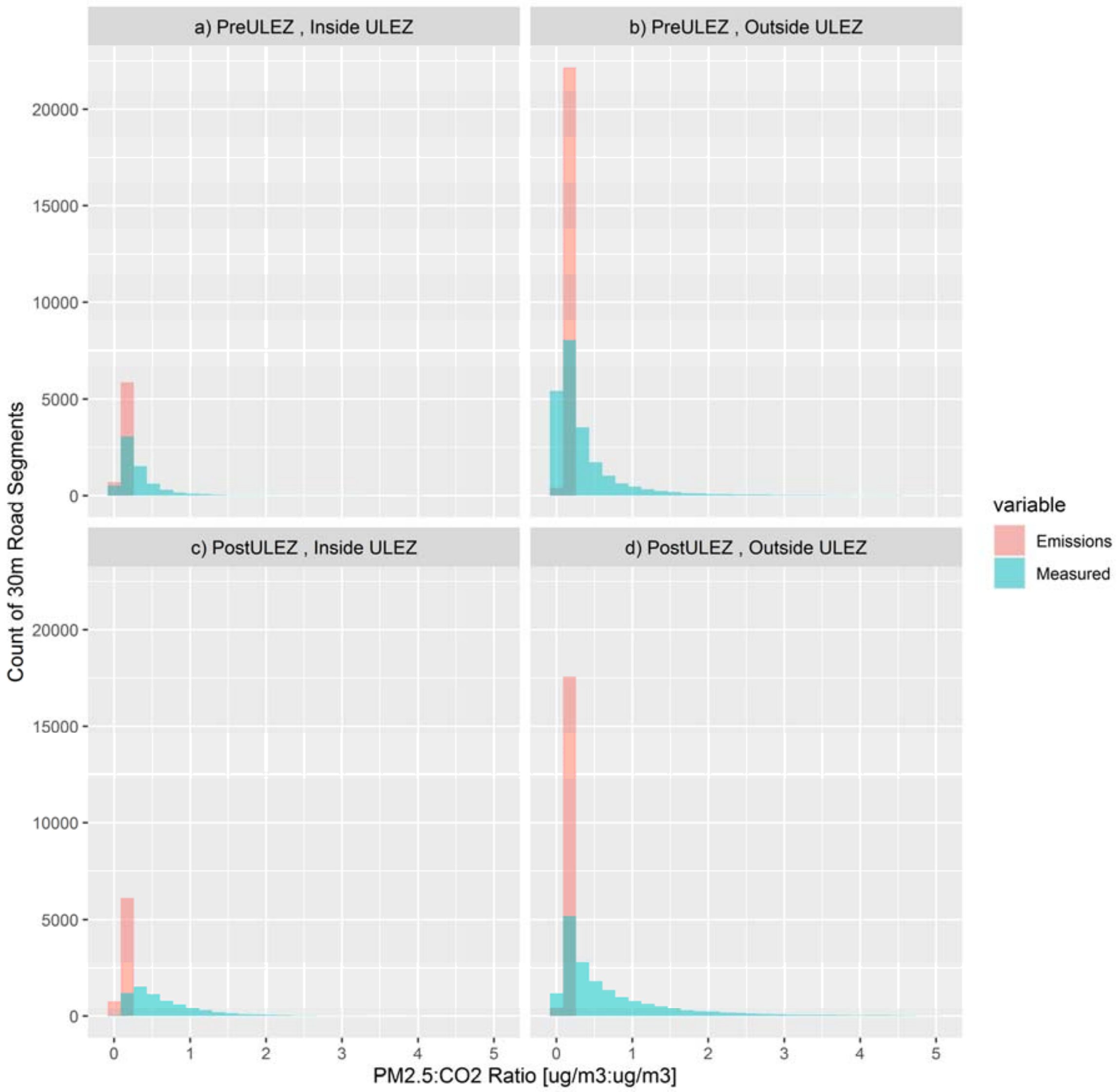

Figure 3-4: Histogram of PM2.5:CO2 (ugm3:ppm) ratios in emissions and measured data per 30m road segment, separated by before and after the introduction of the ULEZ and whether the road segment was inside or outside the ULEZ. This dataset has been capped at a ratio value of 5.

[4]Carslaw, D and Rhys-Tyler, G 2013: New insights from comprehensive on-road measurements of NOx, NO2 and NH3 from vehicle emission remote sensing in London, UK. Atmos. Env. 81 pp 339–347.

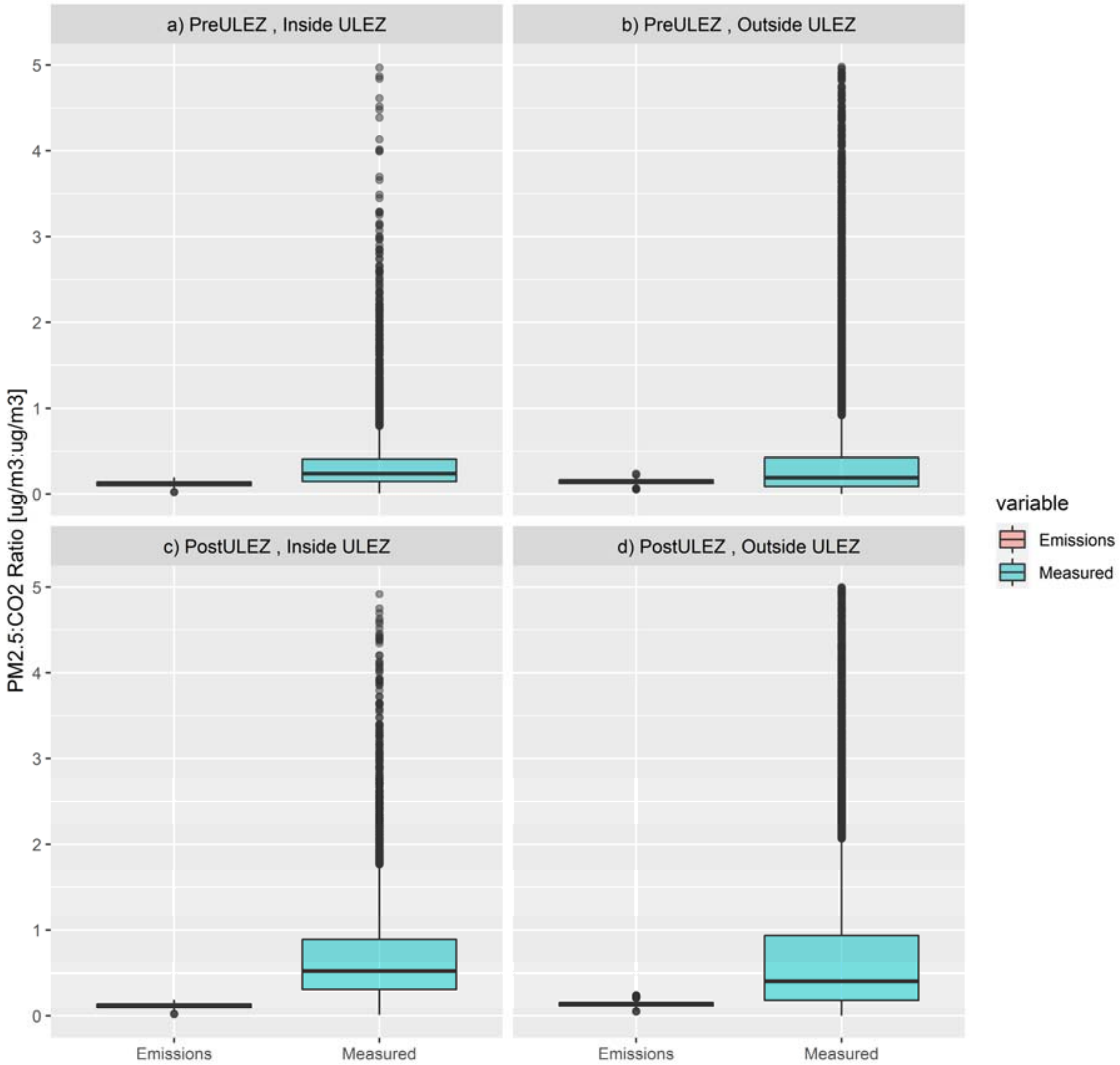

Figure 3-5: Boxplot of PM2.5:CO2 (ugm3:ppm) ratios in emissions and measured data per 30m road segment, separated before and after the introduction of the ULEZ, and whether the road segment was inside or outside the ULEZ. The black dots represent the outliers. This dataset has been capped at a ratio of 5.

Table 3-2 Table of the median PM2.5:CO2 (ugm3:ppm) ratio in emissions and measured data per 30m road segment

		Emissions	Measured
Pre ULEZ	Inside ULEZ	0.12	0.24
Pre ULEZ	Outside ULEZ	0.14	0.19
Post ULEZ	Inside ULEZ	0.12	0.52
Post ULEZ	Outside ULEZ	0.14	0.40

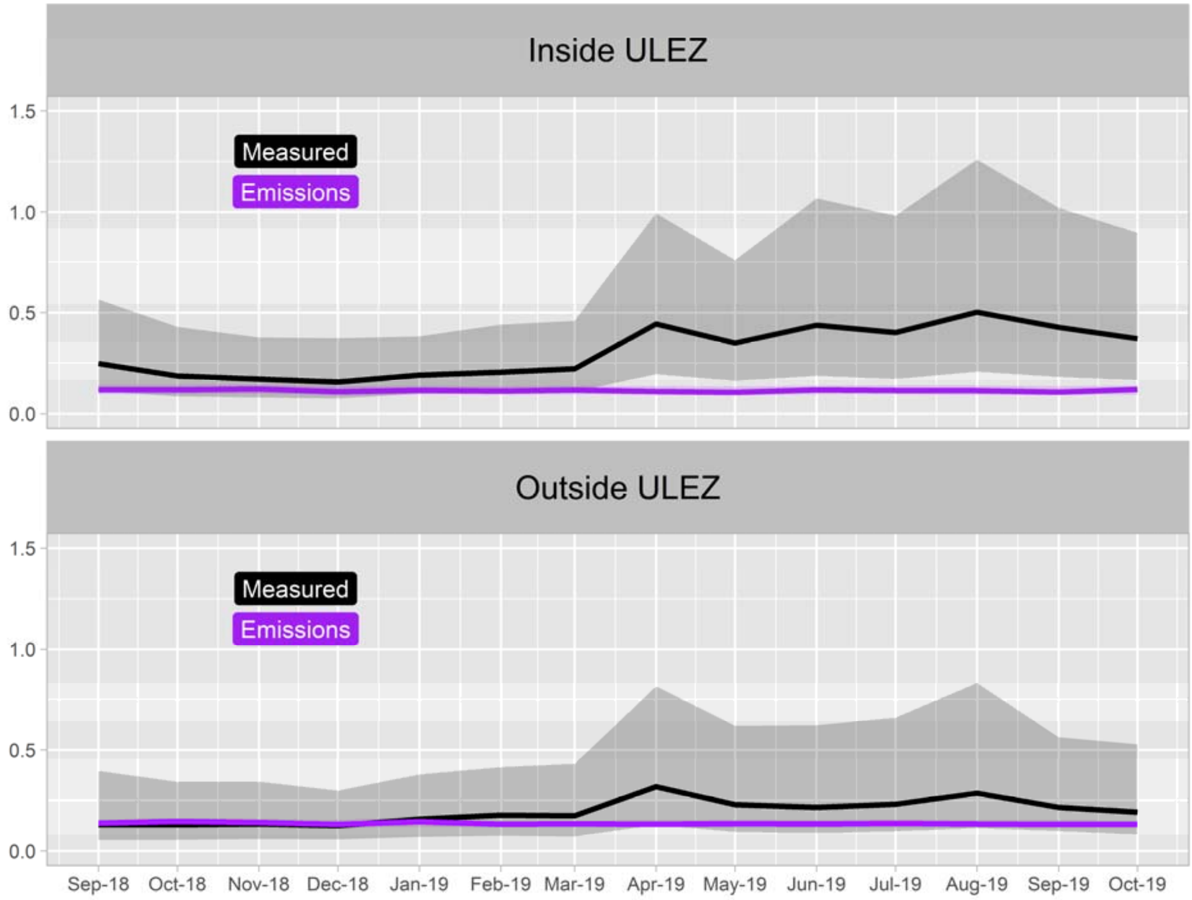

Figure 3-6: A time series of monthly averaged PM2.5:CO2 (ugm3:ppm) ratios in measured data (black) and emissions (purple) for the period from the 1st September 2018 to 31st October 2019, separated by roads inside and outside the ULEZ. The bold line represents the median ratio and the faded lines represent the inter-quartile range

4. Discussion

Overall, the NO_X: CO_2 ratios calculated from the Breathe London mobile measurements are similar to the equivalent ratios calculated from LAEI-based emissions inventory. As expected, the mobile data ratios have a far greater range of values than the emissions inventory ratios because the high frequency mobile data captures the very localised variations in traffic conditions during the drives, whereas the emissions inventory represents the average traffic conditions on each road. There is a noticeable decrease in the monthly median ratio in the measured data from September 2018 through April 2019, before plateauing from April 2019 to October 2019. This suggests an increase in the proportion of cleaner vehicles across London during the period leading up to the introduction of the ULEZ, which is likely to be linked to the introduction of the ULEZ and an increasing vehicle compliance level in the months before the official start. The measured ratio changes outside the ULEZ are likely to be caused by the general uptake of newer, cleaner vehicles over this time, in addition to impacts of the ULEZ on the vehicle fleet outside the ULEZ area.

On the other hand, the $PM_{2.5}$:CO_2 ratios calculated from the Breathe London mobile measurements are higher than the equivalent ratios calculated from LAEI emissions inventory, indicating that adjustments for real world conditions are necessary for $PM_{2.5}$ as well as for NO_X. $PM_{2.5}$ is largely dominated by non-exhaust emissions, and is unlikely to be as influenced by the introduction of the ULEZ in April 2019 as NO_X. This is evidenced in Figure 3-6, where the monthly median ratios from the measured values actually increase after

April 2019, and stay relatively consistent beforehand, with the effect being more pronounced inside the ULEZ than outside. This increase is more likely to be caused by a decrease in CO_2 emissions associated with a newer cleaner vehicle fleet in response to the introduction of the ULEZ rather than an increase in $PM_{2.5}$ emissions. Future investigations may be done to analyse the impact of applying real world adjustments to the $PM_{2.5}$ emissions on the emissions inventory ratios.

Additional analysis at the Breathe London static monitoring sites was not possible in this investigation due to the lack of calibrated CO_2 measurements from the AQMesh sensors. For the analysis in this report, only the mobile measurements from the Google Cars were used; these were compared directly with emissions because dispersion and atmospheric chemistry effects are less important where the mobile measurements were captured, along the middle of the road very close to the source of emissions. However, if a similar analysis were to be carried out using AQMesh data, it would be important to account for these effects because AQMesh sensors are located further away from the sources. In that case, pollutant:CO_2 ratios calculated from ADMS modelled concentrations of NO_X, $PM_{2.5}$ and CO_2 at the AQMesh sites would be needed in place of the ratios calculated from emissions in this analysis. A baseline for each hour for each site for each pollutant would need to be subtracted from both modelled and measured values to leave only the hyperlocal enhancement component of each for the calculation of ratios to be meaningful. This hourly baseline could be derived on a site-by-site basis by taking the 5^{th} percentile of the 1-minute measurements for each hour, site and pollutant. Pollutant:CO_2 ratios calculated from AQMesh data in this way would provide a greater level of uncertainty in the interpretation of the results than ratios calculated from mobile data, due to the increasing importance of dispersion and chemistry effects with increasing distance from the source.